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NORMAL-STATE ELECTRONIC PROPERTIES OF $Sn_{0.12}Eu_{1.08}Mo_6S_8$ AT LOW TEMPERATURE AND HIGH PRESSURE

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The Hall coefficient and resistivity of $Sn_{0.12}Eu_{1.08}Mo_6S_8$ have been measured over the range $1.5<T<300$ K under hydrostatic pressure up to 5.3 kbar. The data suggest thermally-activated conduction, but in all cases the carrier concentration saturates at $\sim 10^{22}$ holes/cm^3 for $T>100$ K. A band model is proposed which is qualitatively consistent with the data.

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The Chevrel-phase compound $Eu_{1.2}Mo_6S_8$ exhibits unusual electronic properties at low temperatures. The ambient-pressure resistivity (ρ) is nearly temperature-independent above 100 K but shows a semiconductor-like increase by a factor of $\sim$7 as the temperature is lowered to 4 K [1]. No superconductivity was observed down to 17 mK [2]. However, application of hydrostatic pressure or substitution of Sn for Eu to form $Sn_xEu_{1.2-x}Mo_6S_8$ was found to induce superconductivity while reducing the normal-state resistivity [3,4]. This gives rise to a regime in which relatively-high-T_c superconductivity (onset temperature as large as 11 K) is observed in samples in which $\partial\rho/\partial T<0$ above T_c [3-5]. Hall-effect measurements on such samples at ambient pressure [4] indicated carrier concentrations (n) as small as 10^{19} cm^{-3} at 12 K. T_c is therefore considerably higher than found in typical low-n superconductors. Further increases in pressure or Sn concentration give rise to metallic-like resistivity behavior in the normal state with $T_c\sim$11 K [3-5].

In this paper we report Hall coefficient (R_H) and resistivity measurements on $Sn_{0.12}Eu_{1.08}Mo_6S_8$ over the range $1.5<T<300$ K under hydrostatic pressures up to 5.3 kbar. In this pressure range the onset temperature for superconductivity shifts with pressure from below our lowest temperature to $\sim$9 K while $\partial\rho/\partial T$ remains negative at $T>T_c$. We therefore obtain information on the electronic structure in this interesting regime. We then present a semiconductor model which is qualitatively consistent with the data.

Samples were prepared as described in ref. 4. Chemical analysis showed the composition to be $Sn_{0.09}Eu_{1.1}Mo_6S_{7.6}$. The sulfur deficiency is typical of Eu-Mo-S samples. Pressure was generated by the self-clamp method [6] and measured by a superconducting Sn manometer placed near the sample. ρ and R_H were measured by standard techniques using low-frequency

($f \sim 8$ Hz) alternating currents and static magnetic fields of 2-12 kG. On the basis of magnetization and sample-density measurements, we conclude that errors in R_H due to the anomalous Hall effect and porosity of the samples are small.

ρ is plotted vs T in Fig. 1. The influence of pressure on the normal-state resistivity is clearly visible. The data at 5.3 kbar show a drop in ρ beginning at ~ 9 K which appears to be the onset of superconductivity.

Carrier concentration (n) derived from the one-band formula $n \equiv (e|R_H|)^{-1}$ is plotted vs T in Fig. 2. The sign reversal of R_H at low pressures indicates that two or more bands of carriers contribute to the Hall voltage. However, if the Hall voltage is assumed to arise from just two bands, it can be shown [7] that the number derived from the one-band formula is an upper limit on the concentration of carriers whose sign is the same as that of R_H. At temperatures rather far removed from the sign reversal, n therefore provides an estimate of the carrier concentration in one of the bands.

It is generally believed that the superconductivity of the Chevrel-phase compounds involves electrons in an incompletely-filled Mo d-subband. In a recent band calculation on Eu-Mo-S [8], it was found that the Fermi level lies in such a band which is located below a sizable forbidden gap (~ 1 eV). A model which rationalizes the main features of the present data and the lack of superconductivity in Eu-Mo-S at ambient pressure involves the introduction of an impurity band containing a fairly large number of states located in the gap on the order of 10 meV above the Mo band. The impurity states could be associated with the significant sulfur deficiency, europium excess, and/or other defects that tend to occur in these compounds. For example, the sulfur deficiency may lead to a filling of the Mo states; and the data indicate that at low temperatures the Fermi level lies near the bottom of the impurity band rather than in the Mo band. Previous resistivity data [3,4] also indicate that for $P \gtrsim 10$ kbar the two bands merge (corresponding to a reasonable pressure coefficient). The model is then consistent with our ($P \lesssim 5$ kbar) data in that: (1) $\partial\rho/\partial T < 0$, $\partial n/\partial T > 0$; (2) there is multi-band behavior, and the temperature at which R_H changes sign decreases with increasing pressure; and (3) n saturates at high temperatures. The behavior at the pressure where the bands begin to overlap is expected to be complex. At higher pressures ($\gtrsim 10$ kbar) the model suggests that $\partial\rho/\partial T$ would be positive as previously observed [3,4] and that n would be relatively independent of temperature.

Measurements at higher pressures in $Eu_{1.2}Mo_6S_8$ and other compositions are currently in progress.

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FIGURE CAPTIONS

Figure 1 : Resistivity vs temperature

Figure 2 : Carrier concentration as defined by $n \equiv (e|R_H|)^{-1}$ vs temperature

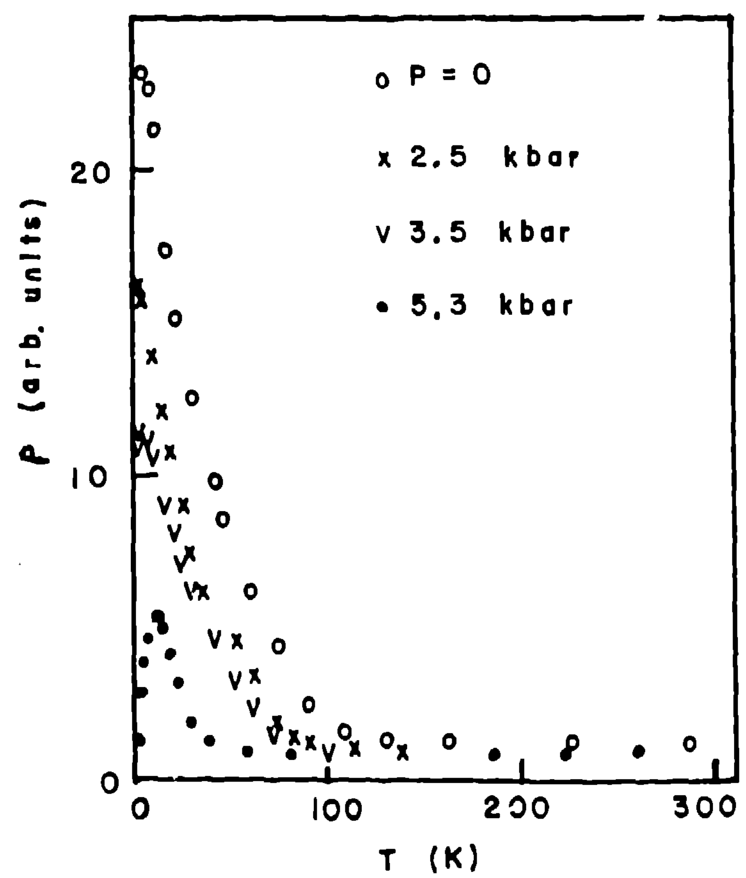

FIGURE 1

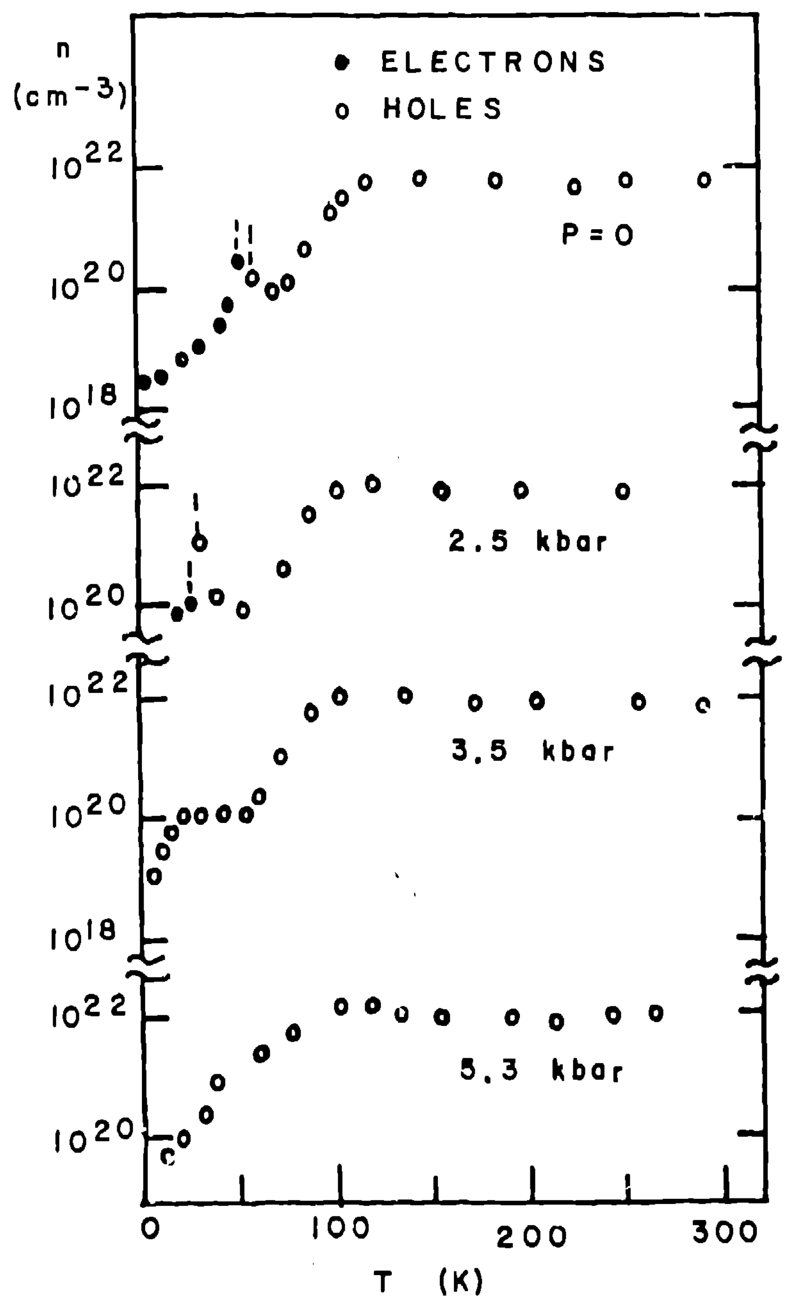
n
(cm-3)
ELECTRONS
HOLES
10²²
10²⁰
10¹⁸
P = 0
2.5 kbar
3.5 kbar
5.3 kbar
0
100
200
300
T (K)

FIGURE 2